

THERMODYNAMICS OF A PSEUDOSPIN-ELECTRON MODEL

Tabunshchyk K.V.

*Institute for Condensed Matter Physics Nat. Acad. Sci. Ukr.
1 Svientsitskii Str., UA-79011 Lviv, Ukraine*

Abstract

The purpose of this article is to present the thermodynamics of the pseudospin electron (PE) model in the case of the different type interactions between pseudospins. First, we provide an overview of the results of works which deal with the theoretical investigation of the PE model with the inclusion of the direct pseudospin-pseudospin interaction (but without the electron transfer). Second, we present the results of the investigation of the model in the case of the absence of the direct pseudospin-pseudospin interaction and Hubbard correlation, when interaction between pseudospins via conducting electron is done.

Introduction

Pseudospin-electron (PE) model is one of theoretical models which considers the interaction of electrons with local lattice vibrations where an anharmonic variables are represented by pseudospins. The theoretical investigation of the PE model is an enduring subject of interest at the quantum statistics department.

The model is used to describe the strongly correlated electrons of CuO_2 sheets coupled with the vibrational states of apex oxygen ions O_{IV} (which move in the double-well potential) in YBaCuO type high- T_c superconductors (HTSC) [1]. Recently a similar model has been applied for investigation of the proton-electron interaction in molecular and crystalline systems with hydrogen bonds [2].

The model Hamiltonian is the following:

$$H = \sum_i H_i + \sum_{ij\sigma} t_{ij} a_{i\sigma}^+ a_{j\sigma} - \frac{1}{2} \sum_{ij} J_{ij} S_i^z S_j^z, \quad (1)$$

$$H_i = U n_{i\uparrow} n_{i\downarrow} - \mu \sum_{\sigma} n_{i\sigma} + g \sum_{\sigma} n_{i\sigma} S_i^z - h S_i^z,$$

where the strong single-site electron correlation U , interaction with the anharmonic mode (g -term), and the energy of the anharmonic potential asymmetry (h -term) are included in the single-site part. Hamiltonian (1) also contains terms, which describe electron transfer t_{ij} and direct interaction between pseudospins J_{ij} . The energy of the electron states at the lattice site is accounted from the level of chemical potential μ .

In the case of strong coupling ($g \gg t$) and correlation ($U \gg t$) the perturbation theory can not be applied. It is reasonable to include these one in zero order Hamiltonian (single-site Hamiltonian H_i). Its eigenfunctions are build of the vectors $|n_{i\uparrow}, n_{i\downarrow}, S_i^z\rangle$, which form the full unit cell state basis [3]:

$$|1\rangle = |0, 0, \frac{1}{2}\rangle, \quad |2\rangle = |1, 1, \frac{1}{2}\rangle, \quad |3\rangle = |0, 1, \frac{1}{2}\rangle, \quad |4\rangle = |1, 0, \frac{1}{2}\rangle, \quad (2)$$

$$|\tilde{1}\rangle = |0, 0, -\frac{1}{2}\rangle, \quad |\tilde{2}\rangle = |1, 1, -\frac{1}{2}\rangle, \quad |\tilde{3}\rangle = |0, 1, -\frac{1}{2}\rangle, \quad |\tilde{4}\rangle = |1, 0, -\frac{1}{2}\rangle.$$

In the early works the main attention at the investigation of this model has been paid to the examination of electron states, effective electron-electron interaction, to the elucidation of additional possibilities of occurrence of superconducting pair correlations. On the basis of PE model a possible connection between the superconductivity and lattice instability of the ferroelectric type in HTSC has been discussed [4, 5].

A series of works has been carried out in which the pseudospin $\langle S^z S^z \rangle$, mixed $\langle S^z n \rangle$ and charge $\langle nn \rangle$ correlation functions were calculated. It has been shown with the use of the generalized random phase approximation (GRPA) in the limit $U \rightarrow \infty$ [3, 6], that there exists a possibility of divergences of these functions at some values of temperature. This effect was interpreted as a manifestation of dielectric instability or ferroelectric type anomaly. The tendency to the spatially modulated charge and pseudospin ordering at the certain model parameter values was found out. The analysis of ferroelectric type instabilities in the two-sublattice model of high temperature superconducting systems has been made [7]. The influence of oxygen nonstoichiometry on localization of apex oxygen in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ type crystals is studied in the work [8].

The purpose of this article (lecture) is to present the thermodynamics of the PE model in the case of the different type interactions between pseudospins. First, we provide an overview of the results of works which deal with the theoretical investigation of the PE model with the inclusion of the *direct pseudospin-pseudospin interaction* (but without the electron transfer ($t_{ij} = 0$)). Second, we present the results of the investigation of the model in the case of the absence of the direct pseudospin-pseudospin interaction and Hubbard correlation ($J_{ij} = 0$, $U = 0$), when *interaction between pseudospins via conducting electron is done*.

1. Direct interaction between pseudospins.

1.1. Ferroelectric type interaction.

The work [9] is devoted to the study of the PE model in the case of zero electron transfer ($t_{ij} = 0$). The direct interaction between pseudospins is taken into account. It is supposed to be a long-ranged that allows one to use the mean field approximation (MFA). In this approximation, the model Hamiltonian has the following form:

$$H = \sum_i \tilde{H}_i + \frac{N}{2} J \eta^2, \quad \tilde{H}_i = -\mu \sum_{\sigma} n_{i\sigma} + U n_{i\uparrow} n_{i\downarrow} + g \sum_{\sigma} n_{i\sigma} S_i^z - (h + J\eta) S_i^z.$$

The interaction ($J_{ij} \sim J/N$) is taken as the ferroelectric type one; the order parameter $\eta = \langle S_i^z \rangle$ does not depend on the unit cell index.

Grand canonical potential and partition function of the model, calculated per one lattice site are equal to

$$\begin{aligned} \frac{\Omega}{N} &= -T \ln Z_i + \frac{1}{2} J \eta^2, \\ Z_i &= 2 \left[\cosh \beta h_n + e^{-\beta(U-2\mu)} \cosh \beta(h_n - g) + 2e^{\beta\mu} \cosh \beta \left(h_n - \frac{g}{2} \right) \right], \end{aligned} \quad (3)$$

where $h_n = h/2 + J\eta/2$.

Then, all thermodynamic properties can be derived from the presented formulae (3).

The average number of electrons is determined as follows [9]:

$$-\frac{1}{N} \left(\frac{\partial \Omega}{\partial \mu} \right)_{T,h,\mu} = \left\langle \frac{1}{N} \sum_i n_i \right\rangle \equiv \langle n \rangle. \quad (4)$$

The equation for the order parameter is obtained from the thermodynamical relation [9]:

$$\left(\frac{\partial \Omega}{\partial \eta} \right)_{T,h,\mu} = 0. \quad (5)$$

For the investigation of equilibrium conditions one should separate two regimes: $\mu = \text{const}$ and $n = \text{const}$. We would like to note that hereafter we shall demonstrate the results of numerical investigation which show the main features of the considered model.

1.1.1. $\mu = \text{const}$ regime.

The $\mu = \text{const}$ regime corresponds to the case when, for example, charge redistribution between the conducting sheets CuO_2 and other structural elements is allowed. For this regime the equilibrium is defined by the minimum of the grand canonical potential that form an equation for pseudospin mean value (5) and expression for $\langle n \rangle$ (4).

At some regions of h values the field dependencies $\eta(h)$ possesses S -like behaviour Fig. 1 (the first order phase transition with the jump of order parameter and electron concentration take place at the change of field h). The phase transition point is presented by a crossing point on the dependence $\Omega(h)$.

The phase diagram $T_c - h$ is shown in Fig. 2. One can see that with respect to Ising model the phase coexistence curve is shifted in field and distorted from the vertical line and hence the possibility of the first order phase transition with the temperature change exists in PE model.

The phase diagram $\mu - h$ (Fig. 2) indicate stability regions for states with $\eta = \pm 1/2$ ($U < g$ and $T = 0$). The form of diagram depends on the relation between U and g parameter values [9]. Transitions between regions $|r\rangle \leftrightarrow |p\rangle$, $|\tilde{r}\rangle \leftrightarrow |\tilde{p}\rangle$ lead to the change of the average number of electrons only. At transitions $|r\rangle \leftrightarrow |\tilde{r}\rangle$ the flipping of pseudospin takes place, and at $|r\rangle \leftrightarrow |\tilde{p}\rangle$ ($r \neq p$) both processes occur.

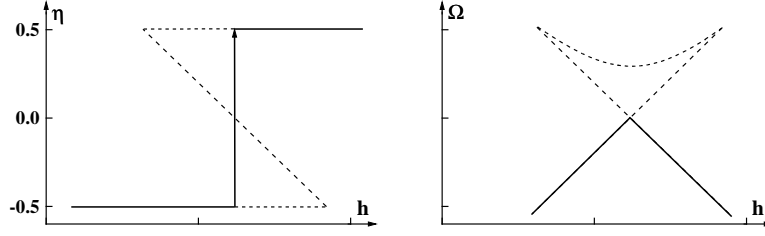


Figure 1. Field dependencies of the order parameter and grand canonical potential ($T = 0$).

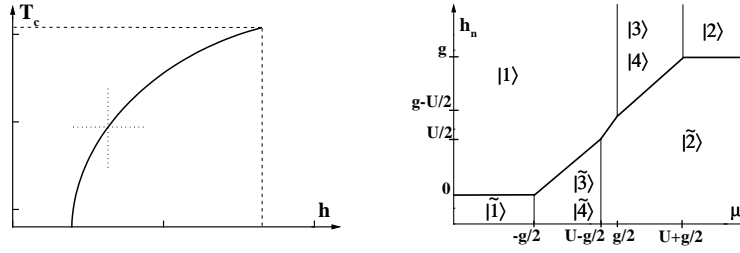


Figure 2. $T_c - h$ and $\mu - h$ phase diagrams.

Hence, the possibility of the first order phase transition with the change of field h and/or chemical potential μ takes place and is shown by thick line on phase diagram in Fig. 2.

1.1.2. $n = \text{const}$ regime.

In the regime of the fixed electron concentration value the equilibrium is determined by the minimum of free energy $F = \Omega + \mu N$ and form a set of equations (4), (5) for the chemical potential and order parameter.

Typical example of the $\eta(h)$ dependence is shown in Fig. 3 which corresponds to the $\langle n \rangle$ value in the interval $0 \leq n \leq 1$. Phases $\eta = -\frac{1}{2}$ (phase 1), $\eta = \frac{1}{2} - n$ (phase 2), $\eta = \frac{1}{2} - \frac{n}{2}$ (phase 3), $\eta = \frac{1}{2}$ (phase 4) exist between phase transition points (which is determined according to the Maxwell rule from the range of S -like behaviour (between the spinodal points Z_i)) and outside of them. At the change of the model parameter values the

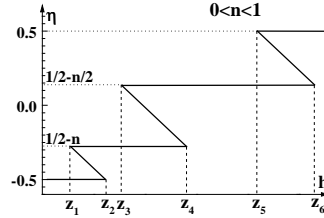


Figure 3. Field dependency of the order parameter ($T = 0$).

regions, where metastable phases exist, can overlap, some phase transitions disappear (some intermediate phases can not be realized). In case $1 \leq n \leq 2$ the dependence $\eta(h)$ is generally similar. The phase 3 and phase 2' at $\eta = \frac{3}{2} - n$, which now appears instead of phase 2, may play the role of the intermediate phases.

On the phase diagram $n - h$ Fig. 4 the thick solid line indicates the phase coexistence curve and hence the possibility of the first order phase transition with the change of the longitudinal field h and/or electron concentration $\langle n \rangle$ takes place.

More detail analyse of a free energy behaviour shows that the above presented (in this paragraph) results are not realized. The investigation of the equilibrium conditions shows that the first order phase transition transforms into the phase separation. One can see regions where state with homogenous distribution of particles is unstable ($d\mu/dn \leq 0$), and the phase separation into the regions with different concentrations exists (Fig. 5). The phase diagram $n - h$ (Fig. 5) illustrates the separation phases.

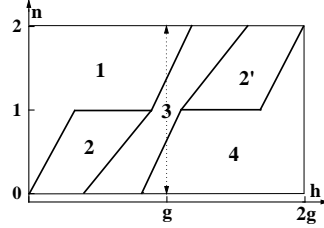


Figure 4. $n - h$ phase transition diagram ($T = 0$).

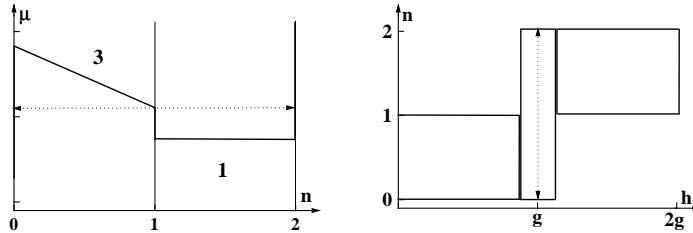


Figure 5. Dependence of the chemical potential μ on the electron concentration and $n - h$ phase separation diagram ($T = 0$).

The phase 3 splits into phase 4 (with concentration $n = 0$, order parameter $\eta = \frac{1}{2}$) and phase 1 ($n = 2$, $\eta = -\frac{1}{2}$) with weight coefficients $1 - n$ and n respectively (thin dotted lines in Fig. 4 and Fig. 5).

Therefore, more convenient and thermodynamically stable is the phase separated state, which is the mixture of states with different electron concentrations and different values of order parameters.

1.2. Antiferroelectric type interaction.

In the case of antiferroelectric type interaction it is convenient to introduce two kinds of sites (A-sites, B-sites). These corresponds to the

doubling of the lattice period [10].

Within the framework of the MFA we shall write:

$$S_A^z S_B^z = -\eta_A \eta_B + \eta_A S_B^z + \eta_B S_A^z, \quad (6)$$

where $\eta_A = \langle S_A^z \rangle$, $\eta_B = \langle S_B^z \rangle$.

Then, we obtain the following expression for the model Hamiltonian:

$$\begin{aligned} H &= \sum_i \tilde{H}_{iA} + \sum_j \tilde{H}_{jB} + N \left\{ \frac{J_1}{2} \eta_A \eta_B + \frac{J_2}{4} (\eta_A^2 + \eta_B^2) \right\}, \\ \tilde{H}_{iA} &= -\mu(n_{iA\downarrow} + n_{iA\uparrow}) + U n_{iA\downarrow} n_{iA\uparrow} + g(n_{iA\downarrow} + n_{iA\uparrow}) S_{iA}^z - \\ &\quad -(h + J_1 \eta_B + J_2 \eta_A) S_{iA}^z, \\ \tilde{H}_{jB} &= \tilde{H}_{jA}|_{A \leftrightarrow B}. \end{aligned} \quad (7)$$

The Hilbert space forms as a direct product of the eigenfunctions (2) for \tilde{H}_A and \tilde{H}_B operators (7) $\{|n_{iA\uparrow} N_{iA\downarrow}, S_{iA}^z\rangle\} \oplus \{|n_{iB\uparrow} N_{iB\downarrow}, S_{iB}^z\rangle\}$. The analytical consideration in this case in general is very similar to the previous (ferromagnetic interaction) one, but formulae are more complicated (see in details [10]).

Grand canonical potential can be written in the form:

$$\begin{aligned} \Omega &= J_1 \eta_A \eta_B + \frac{J_2}{2} (\eta_A^2 + \eta_B^2) + T \ln \left\{ \left(\eta_A^2 - \frac{1}{2} \right) \left(\eta_B^2 - \frac{1}{2} \right) \right\} \\ &\quad - T \ln \left(1 + e^{-\beta(-2\mu+U+g)} + 2e^{-\beta(-\mu+\frac{g}{2})} \right) \\ &\quad - T \ln \left(1 + e^{-\beta(-2\mu+U-g)} + 2e^{-\beta(-\mu-\frac{g}{2})} \right). \end{aligned} \quad (8)$$

1.2.1. $\mu = \text{const}$ regime.

The set of equations for η_A , η_B is defined by the minimum of the grand canonical potential (8). The expression for the electron mean values is determined from the thermodynamical relation (4).

The form of the grand canonical potential field dependence $\Omega(h)$ (and therefore the type and number of phase transitions) depends on the relation between parameters J_1 and J_2 values (Fig. 6). There are no any specific behaviour when J_1 and J_2 are placed in the regions 5 and 6. The case when J_1 and J_2 are placed in the domains 1, 7, 8 is similar to ferroelectric type interaction.

The location of J_1 and J_2 parameters within the area 4 leads to the possibility of the two sequential second order phase transitions: from

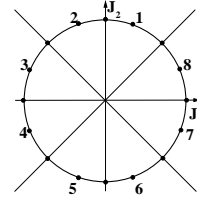


Figure 6. J_1 - J_2 area.

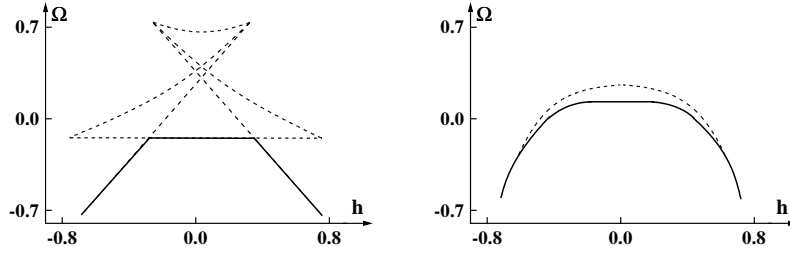


Figure 7. Field dependencies of grand canonical potential within the 2 and 4 regions respectively ($T = 0$).

the ferroelectric phase (FP) with the one pseudospin mean value to the antiferroelectric phase (AP) and then to the FP with the another order parameter value (Fig. 7).

The case when J_1 and J_2 belong to the region 2, 3 is shown in Fig. 7 and the corresponding phase diagram μ - h in Fig. 8.

One can see that two first order phase transitions with the change of the field h and/or chemical potential μ take place. With respect to ferroelectric type interaction between pseudospins (Fig. 2) the phase coexistence curve is split and one obtains the range (the range width is equal J_1) where the AP exists (Fig. 8). With the temperature increase the first order phase transitions transform into the second order phase transitions. Hence the possibility of the first order phase transition from FP into AP and then the second order phase transition from AP into FP exist with the temperature increase for the narrow range of h values.

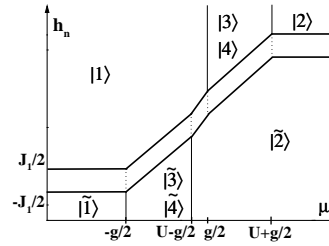


Figure 8. μ - h phase diagram ($T = 0$).

1.2.2. $n = \text{const}$ regime.

As it was mentioned above, in the case of the fixed value of the electron concentration (regime $n = \text{const}$) the first order phase transition transform into the phase separation.

In Fig. 9 one present the phase diagram n - h when J_1 , J_2 are placed in the region 2. Within the area surrounded by the lines the phase separation into the regions with different concentrations and phases FP (solid lines) and AP (dotted lines) takes place. Outside of these boundaries (which surround the phase separated states) the state with the

space homogeneity of electron concentration (FP) is stable. Between the boundaries one have the AP.

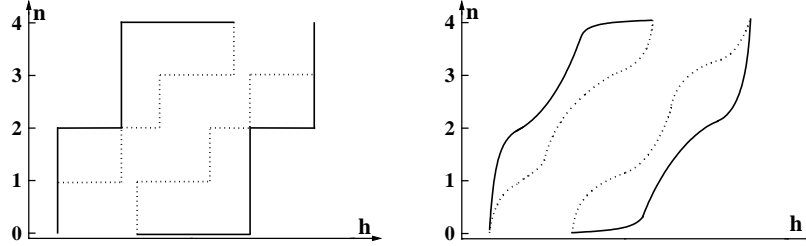


Figure 9. $n - h$ phase diagram ($T = 0$ and $T \neq 0$).

We would like to remind that in the $\mu = \text{const}$ regime with the temperature increase the first order phase transitions transform into the second order ($J_1, J_2 \in \text{domain } 2,3$). On the other hand, in the $n = \text{const}$ regime this correspond to the narrowing of the range of the phase separated states and transform into the second order phase transition curves. Then the phase transition curves approach one to another and, finally, disappear at the certain value of temperature.

The location of J_1, J_2 within the area 4 leads to the possibility of the two second order phase transitions with the change of the field (similar to the $\mu = \text{const}$ regime).

2. Interaction between pseudospins via conducting electron.

In the $U = 0$ and $J_{ij} = 0$ limit operator (1) can be transformed to the Hamiltonian of the electron subsystem of binary alloy in the case of equilibrium disorder. Model (1) is close to the Falicov-Kimball (FK) model but differ in thermodynamic equilibrium conditions, i.e. in a way how self-consistency is achieved ($\langle S^z \rangle = \text{const}$ for the FK model and $h = \text{const}$ for the PE one).

In the present part of work we propose (for the case of the $U = 0$ and $J_{ij} = 0$ limit) the self-consistent scheme for calculation of mean values of pseudospin and particle number operators, grand canonical potential as well as correlation functions. The approach is based on the GRPA with the inclusion of the mean field corrections. The possibilities of the phase separation and chess-board phase appearance are investigated [11].

The calculation is performed in the strong coupling case ($g \gg t$) using of single-site states as the basic one. The formalism of electron creation

(annihilation) operators $a_{i\sigma} = b_{i\sigma}P_i^+$, $\tilde{a}_{i\sigma} = b_{i\sigma}P_i^-$ ($P_i^\pm = \frac{1}{2} \pm S_i^z$) acting at a site with the certain pseudospin orientation is introduced. Expansion of the calculated quantities in terms of electron transfer leads to the infinite series of terms containing the averages of the T -products of the $a_{i\sigma}$, $\tilde{a}_{i\sigma}$ operators. The evaluation of such averages is made using the corresponding Wick's theorem. The averages of the products of the projection operators P_i^\pm are expanded in semi-invariants [11].

Nonperturbated electron Green function is equal to

$$g(\omega_n) = \langle g_i(\omega_n) \rangle; \quad g_i(\omega_n) = \frac{P_i^+}{i\omega_n - \varepsilon} + \frac{P_i^-}{i\omega_n - \tilde{\varepsilon}}, \quad (9)$$

where $\varepsilon = -\mu + g/2$, $\tilde{\varepsilon} = -\mu - g/2$ are single-site energies. Single-electron Green function (calculated in Hubbard-I type approximation) is $\Rightarrow = G_{\mathbf{k}}(\omega_n) = (g^{-1}(\omega_n) - t_{\mathbf{k}})^{-1}$ and its poles determine the electron spectrum

$$\varepsilon_{\text{I,II}}(t_{\mathbf{k}}) = \frac{1}{2}(2E_0 + t_{\mathbf{k}}) \pm \frac{1}{2}\sqrt{g^2 + 4t_{\mathbf{k}}\langle S^z \rangle g + t_{\mathbf{k}}^2}. \quad (10)$$

In the adopted approximation the diagrammatic series for the pseudospin mean value can be presented in the form

$$\langle S^z \rangle = \bullet = \bigcirc - \text{loop} + \frac{1}{2!} \text{2-loop} - \dots \quad (11)$$

Here we use the following diagrammatic notations: $\bigcirc - S^z$, $\text{---}\bigcirc\text{---} - g_i(\omega_n)$, wavy line is the Fourier transform of hopping $t_{\mathbf{k}}$. Semi-invariants are represented by ovals and contain the δ -symbols on site indexes. In the spirit of the traditional mean field approach [11] the renormalization of the basic semi-invariant by the insertion of independent loop fragments is taken into account in (11).

The analytical expression for the loop is the following:

$$\begin{aligned} \text{loop} &= \frac{2}{N} \sum_{n,\mathbf{k}} t_{\mathbf{k}}^2 \frac{(P_i^+(i\omega_n - \varepsilon)^{-1} + P_i^-(i\omega_n - \tilde{\varepsilon})^{-1})}{g^{-1}(\omega_n) - t_{\mathbf{k}}} \\ &= \beta(\alpha_1 P_i^+ + \alpha_2 P_i^-). \end{aligned} \quad (12)$$

It should be noted that within the self-consistent scheme of the GRPA, the chain fragments form the single-electron Green function in the Hubbard-I approximation and in the sequences of loop diagrams in

the expressions for grand canonical potential Ω and pair correlation functions ($\langle S_i^z S_j^z \rangle$, $\langle S_i^z n_j \rangle$, $\langle n_i n_j \rangle$) the connections between any two loops by more than one semi-invariant are omitted. This procedure includes the renormalization of the higher order semi-invariants, which is similar to the one given by expression (11).

From (11) and (12) follows the equation for pseudospin mean value

$$\langle S^z \rangle = \frac{1}{2} \tanh \left\{ \frac{\beta}{2} (h + \alpha_2 (\langle S^z \rangle) - \alpha_1 (\langle S^z \rangle)) + \ln \frac{1 + e^{-\beta \varepsilon}}{1 + e^{-\beta \bar{\varepsilon}}} \right\}. \quad (13)$$

The grand canonical potential in the considered approximation has the form:

$$\begin{aligned} \Delta \Omega = \Omega - \Omega \Big|_{t=0} = & -\frac{2}{N\beta} \sum_{\mathbf{k}} \ln \frac{(\cosh \frac{\beta}{2} \varepsilon_I(t_{\mathbf{k}})) (\cosh \frac{\beta}{2} \varepsilon_{II}(t_{\mathbf{k}}))}{(\cosh \frac{\beta}{2} \varepsilon) (\cosh \frac{\beta}{2} \bar{\varepsilon})} + \langle S^z \rangle (\alpha_2 - \alpha_1) \\ & - \frac{1}{\beta} \ln \cosh \left\{ \frac{\beta}{2} (h + \alpha_2 - \alpha_1) + \ln \frac{1 + e^{-\beta \varepsilon}}{1 + e^{-\beta \bar{\varepsilon}}} \right\} + \frac{1}{\beta} \ln \cosh \left\{ \frac{\beta}{2} h + \ln \frac{1 + e^{-\beta \varepsilon}}{1 + e^{-\beta \bar{\varepsilon}}} \right\} \end{aligned}$$

The solution of equation (13) and calculation of potential Ω were performed numerically.

In the $\mu = \text{const}$ regime (in the uniform case) there exists a possibility of the first order phase transition with the jump of the pseudospin mean value and reconstruction of the electron spectrum [11].

In the $n = \text{const}$ regime one can see (Fig. 10) that the regions with $d\mu/dn \leq 0$, where states with a homogeneous distribution of particles are unstable, exist. This corresponds to the phase separation into the states with different electron concentrations and pseudospin mean values. In the phase separated region the free energy as a function of n deflects up (Fig. 10) and concentrations of the separated phases are determined by the tangent line touch points (these points are also the points of binodal lines which are determined according to the Maxwell rule from the function $\mu(n)$).

The analysis of the $\langle S^z S^z \rangle_{\mathbf{q}}$ correlator temperature behaviour shows that the high temperature phase become unstable with respect to fluctuations with $\mathbf{q} \neq 0$ for certain values of model parameters. The maximal temperature of instability is achieved for $\mathbf{q} = (\pi, \pi)$ (in the case of square lattice with nearest-neighbor hopping) and indicates the possibility of phase transition into a modulated (chess-board) phase.

The analytical consideration of the chess-board phase within the framework of the presented above approximation can be performed in a similar way.

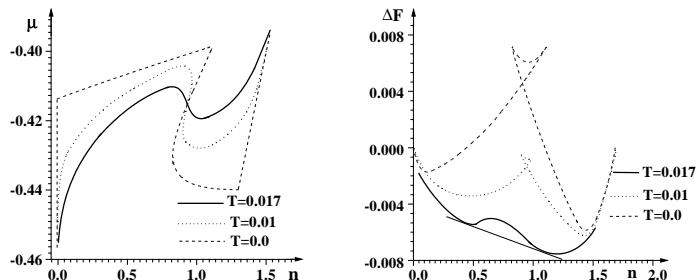


Figure 10. Dependence of the chemical potential μ on the electron concentration n and deviation of the free energy from linear dependence $\Delta F = F(n) - [\frac{n}{2}F(2) + (1 - \frac{n}{2})F(0)]$ for different T values.

From the comparison of the grand canonical potential Ω values for uniform and chess-board phases, the (μ, h) phase diagram is obtained (Fig. 11). One can see that chess-board phase exists as an intermediate one between the uniform phases with the different $\langle S^z \rangle$ and n values. The transition between different uniform phases (bistability) is of the first order (Fig. 11, dashed line), while the transition between the uniform and modulated ones is of the first (dotted line) or second (solid line) order.

Appearance of various phases in the considered model remind the situation known for the FK model with a rich phase diagram [12]. However, contrary to this model, an existence of the phase transitions between uniform phases is possible in our case. This results from the another regime of thermodynamic averaging (fixation of h field which is an analogous to the chemical potential in the FK model).

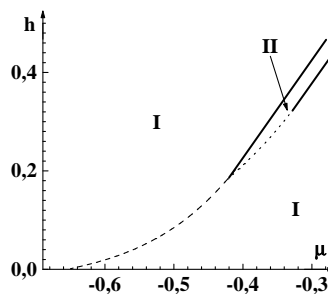


Figure 11. μ - h phase diagram. I – uniform phase, II – chess-board phase.

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Tabunshchik K.V. e-mail: tkir@icmp.lviv.ua